Femtosecond Pulses of Synchrotron Radiation

Laser Time-Slicing Will Lead to Ultrafast Time Resolution

Scientists from Berkeley Lab have generated 300-femtosecond pulses of bend-magnet synchrotron radiation at the Advanced Light Source (ALS) with the aid of a laser "time-slicing" technique. Their proof-of-principle experiment demonstrates that this technique is a viable one for producing ultrashort pulses of x rays. An ALS bend-magnet beamline is already under construction that will be dedicated to time-resolved x-ray diffraction, EXAFS, and other techniques capable of probing the long-range and local structure of matter on a femtosecond time scale.

Atomic motion on the fundamental time scale of a vibrational period (about100 fs), via the making and breaking of chemical bonds and the rearrangement of atoms, ultimately determines the course of phase transitions in solids, the kinetic pathways of chemical reactions, and even the efficiency and function of biological processes. A thorough understanding of such dynamic behavior is a first step to being able to control structural evolution, and it is expected to have important scientific applications in solid-state physics, chemistry, materials science, and biology.

X rays can provide the requisite structural information, and ultrafast x-ray science is an emerging field of research in which x-ray techniques are used in combination with femtosecond lasers to probe structural dynamics. However, the tremendous potential scientific impact of this research area is so far largely unfulfilled, owing to the lack of adequate sources. For example, the pulse length of a synchrotron x-ray source is limited by the bunch length of the electrons in the storage ring, around 30 ps at the ALS.

In early 1996, Alexander Zholents and Max Zolotorev of Berkeley Lab's Center for Beam Physics proposed the laser time-slicing technique as a way to achieve effective bunch lengths in the femtosecond range. At the heart of the proposal was the use of a high-power, femtosecond laser synchronized with the electron bunches so that a pulse of laser light passed collinearly with an electron bunch through an undulator or wiggler. The high electric field of the shorter laser pulse modulated a portion of the longer electron bunch, with some electrons gaining energy and some losing energy. Subsequently, when the energy-modulated electron bunch reached a bend magnet (or other section of the storage ring with a non-zero dispersion), a transverse separation occurred. A collimator or aperture selected the synchrotron radiation from the displaced bunch slices.

To demonstrate the laser bunch-slicing technique, a team led by Robert Schoenlein was established with members drawn from the Berkeley Lab Materials Sciences Division, the Center for Beam Physics, the ALS, and the University of California, Berkeley (UC Berkeley). The team made use of the 16-cm-period wiggler that illuminates Beamline 5.0.2, a test chamber on bend-magnet Beamline 6.3.2), and a high-power titanium-sapphire laser. To verify the femtosecond time structure, they first imaged visible light from Beamline 6.3.2 onto a nonlinear optical crystal along with a delayed 50-fs cross-correlation pulse from the laser system. Then they counted photons at the sum frequency of the two pulses as a function of delay between the modulating and the cross-correlation laser pulses. An adjustable knife edge located in the beamline at an intermediate image plane provided a means to select radiation from different transverse regions of the electron beam. In this way, they measured a dark 300-femtosecond hole in the central cone of the synchrotron radiation and a bright 300-femtosecond peak in the wing of the synchrotron radiation.

As the next step in the growing femtosecond x-ray science program at the ALS under the leadership of Schoenlein and Roger Falcone of UC Berkeley, a bend-magnet beamline (Beamline 5.3.1) is under construction with an anticipated completion date of June 2000. Initial experiments include time-resolved x-ray diffraction, EXAFS, and NEXAFS (XANES).

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R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zholents, and M. S. Zolortorev, "Generation of Femtosecond Pulses of Synchrotron Radiation," *Science* **287**, 2237 (2000).

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FEMTOSECOND PULSES OF SYNCHROTRON RADIATION

Laser Time-Slicing Will Lead to Ultrafast Time Resolution

- **Atomic motion at molecular-vibration time scale (≈100 fs) sets**
  - *Course of phase transitions in solids*
  - *Kinetic pathways of chemical reactions*
  - *Efficiency and function of biological processes*

- **X rays provide direct structural information**
  - *X-ray diffraction for long-range order*
  - *EXAFS, NEXAFS (XANES), SEXAFS for local and surface order*

- **Time resolution slowed by lack of femtosecond x-ray sources**
  - *Pump-probe laser–synchrotron time-resolved experiments*
  - *X-ray pulses limited by bunch length in storage ring to > 30 ps*

- **Laser time-slicing technique demonstrated at the ALS**
  - *300-fs pulses of bend-magnet synchrotron radiation achieved*
  - *New beamline will have x-ray flux and brightness for experiments*

R. W. Schoenlein, S. Chattopadhyay, C. V. Shank, A. A. Zholents, and M. S. Zolotorev (Berkeley Lab); H. H. W. Chong (University of California, Berkeley); and T. E. Glover and P. A. Heimann (ALS). [Science 287 (2000) 2237]

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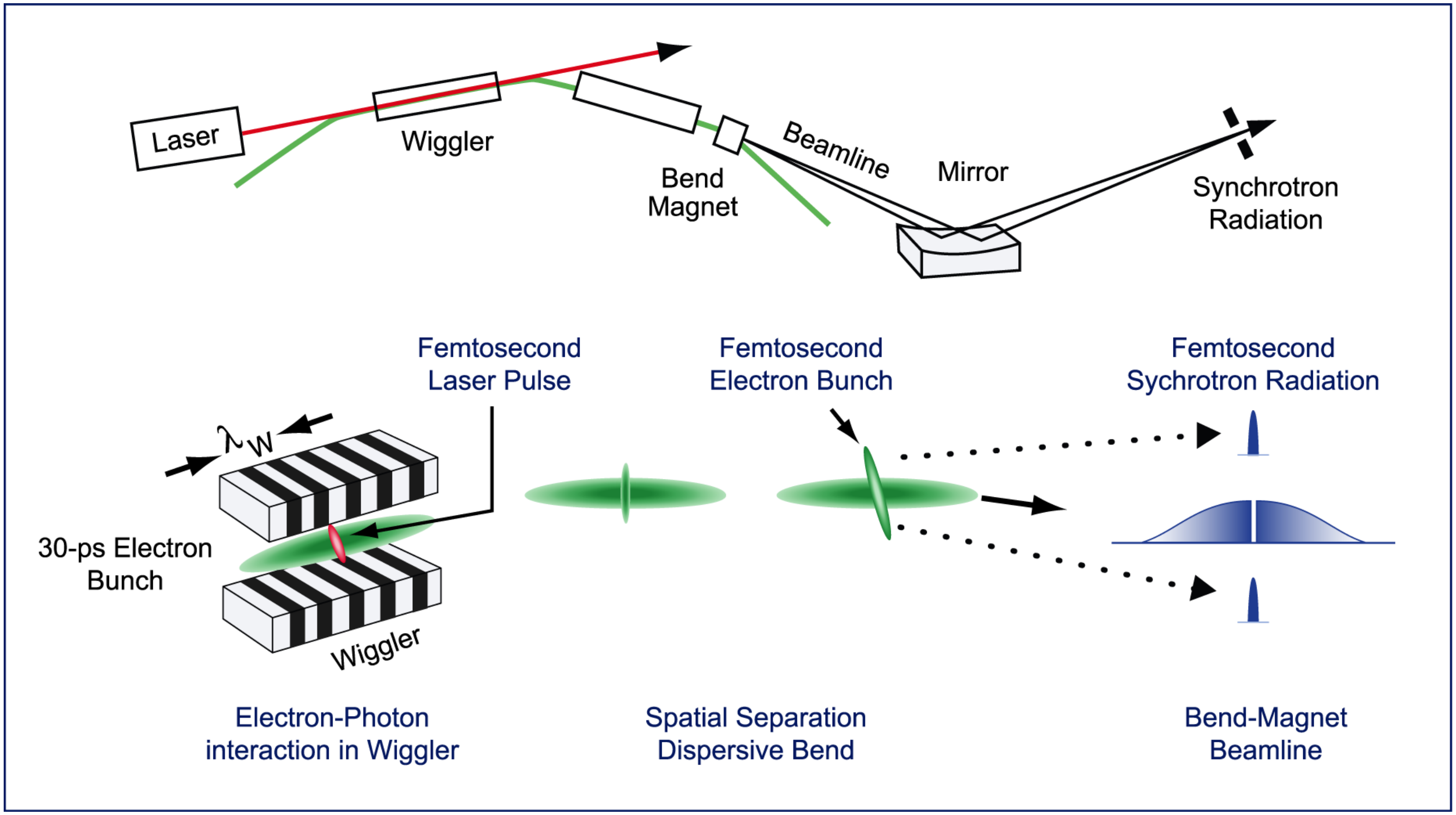

Schematic diagram outlining laser bunch-slicing technique for generating femtosecond pulses of synchrotron radiation at a bend-magnet beamline.

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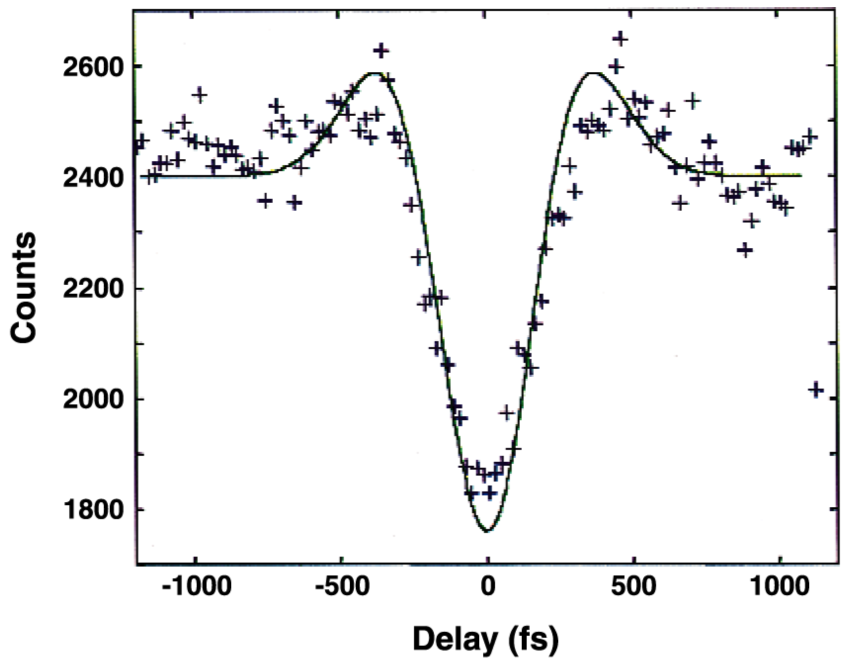
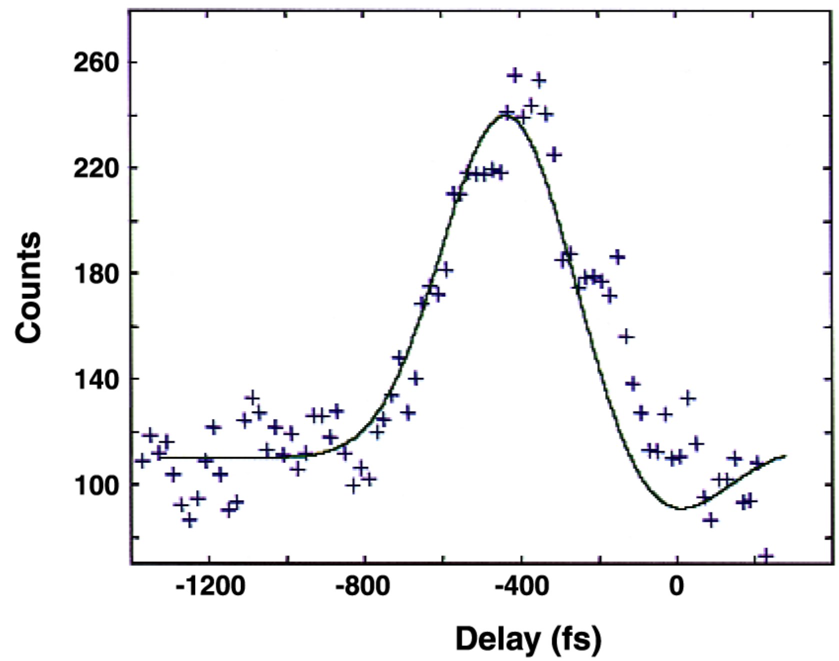


Synchrotron radiation from the central core of the electron bunch shows a dark femtosecond hole.



Synchrotron radiation from a wing of the electron bunch shows a bright femtosecond peak.

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